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1 May 1979 - 30 April 1980

CRITICALLY EVALUATED DATA ON OPTICAL AND TRANSPORT PROPERTIES OF SOLIDS
(Refractive Index and Absorption Coefficient of Selected Optical Materials)

LEVEL II

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By

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Prepared for

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OBJECTIVES

The objectives of this research project is to provide evaluated data and information on optical properties at high temperatures of selected solid materials of potential importance in laser window and laser-hardened material technologies.

The need for ultraviolet and infrared photographic and detecting devices and the development of high-power lasers and their associated applications have resulted in the requirements for a wide variety of highly transparent optical components, such as windows, lenses, and polarizers. In selecting materials of practical importance, one is concerned with insulating or semiconducting solids with wide transparent regions which cover a variety of applications.

Among the various optical properties, those of practical importance are the refractive index and absorption index. The latter is especially important in the application of high-power lasers because many unfavorable effects, which are not observable at low power levels, are developed at high power mainly due to the absorption property of the materials. No matter how low the absorption is, the effect is objectionable at high-power levels.

Over the years, extensive theoretical and experimental investigations have been conducted in an increasing effort to determine the frequency and temperature dependences of the absorption property of optical materials and to identify the mechanisms influencing the absorption. As a result, numerous measurements and calculations have been reported. However, the available information is dispersed throughout the world literature and a comprehensive and exhaustive literature survey and data compilation and analysis is not available. As a result, the majority of users are faced with a dearth of data. It has been often experienced that optical constants data in common usage are often inaccurate or erroneous. For this reason, the current project was activated.

The goals of the present work are:

(i) to review the present state of knowledge on the refractive index absorption coefficient of selected groups of pure solid substances related to the Air Force environment resistant material technology,

(ii) to compile the available data and information,

(iii) to critically evaluate and analyze the existing data, and

(iv) to generate recommended values of these coefficients.

Selection of substances was based on the needs of the Air Force material technology and the recommendation of the National Material Advisory Board on high-power infrared-laser windows. The research program planned to investigate the following groups of substances:

(i) alkali halides

(ii) alkaline earth halides

(iii) diamond-structures materials (Ge and Si)

(iv) (GaP, GaAs, GaSb, InP, InAs and InSb)

(v) (ZnS, ZnSe, ZnTe, CdS, CdSe and CdTe)

(vi) silver halides (AgCl, AgBr and AgI)

(vii) oth[illegible] materials to be selected.

We have completed [illegible] works for material groups (i) and (ii) and are currently (at the time of writing this report) investigating groups (iii) and (iv).

STATUS OF THE RESEARCH EFFORT

In this period we completed the research on the absorption coefficient of alkaline earth halides and a technical report entitled "Absorption Coefficient of Alkaline Earth Halides" was submitted for inspection and acceptance.

In this report, we presented all the available experimental data of absorption coefficient and related information useful for the Air Force material technologies and laser researches. In the data analysis on the available data of laser interest, we have developed an equation, eq. (1), that describes the absorption coefficient as a function of both frequency and temperature.

$$\alpha(\nu,T) = \alpha_0 e^{-a(\nu+b)(c-\log T)} \qquad (1)$$

where α_0, a, b, and c are constants for a given material. These constants, listed in Table 1, were determined based on data evaluation, correlation, and synthesis. In the example of strontium fluoride, Figures 1 and 2 provide a visual comparison of the predicted values of eq. (1) and experimentally determined data. Figure 3 is the corresponding calculated value for a wider region including the crossover point (b,α_0).

Table 1. The Parameters of Equation (1)

Crystal	$\log \alpha_0$	a, cm	b, cm^{-1}	c
MgF_2	3.1877	0.004184	-377.29	4.2901
CaF_2	3.0834	0.005251	-383.92	4.4498
SrF_2	2.9169	0.006577	-360.56	4.3288
BaF_2	2.8507	0.007349	-327.67	4.2873

As a counterpart of the Urbach rule for the uv absorption edge, we have established the expression, eq. (1), for the infrared absorption edge. These expressions are of the same form and the parameters in the corresponding equations are similar. Compared with Deutsch's (eq. 2) expression, we have extended the dependence of the absorption coefficient to include the temperature in addition to frequency.

$$\alpha(\nu) = \alpha_0 e^{-\nu/\nu_0} \qquad (2)$$

It is worthwhile to point out that the equation formulated in this work is of the same type as it was found for alkali halides. Correlation techniques used for alkali halides are equally applicable in the present work.

The work on the absorption coefficient of germanium and silicon was in the stage of data extraction and compilation. A large portion of the available experimental data known to us is dealing with film specimens in the studies of interband and intraband transitions. Large discrepancies and inconsistencies are found among data reported from different sources as the result of that absorption is very sensitive to impurities as well as to the sample preparation procedures. The work of data evaluation will therefore be largely in the characterization of the material specimens and conditions of measurements.

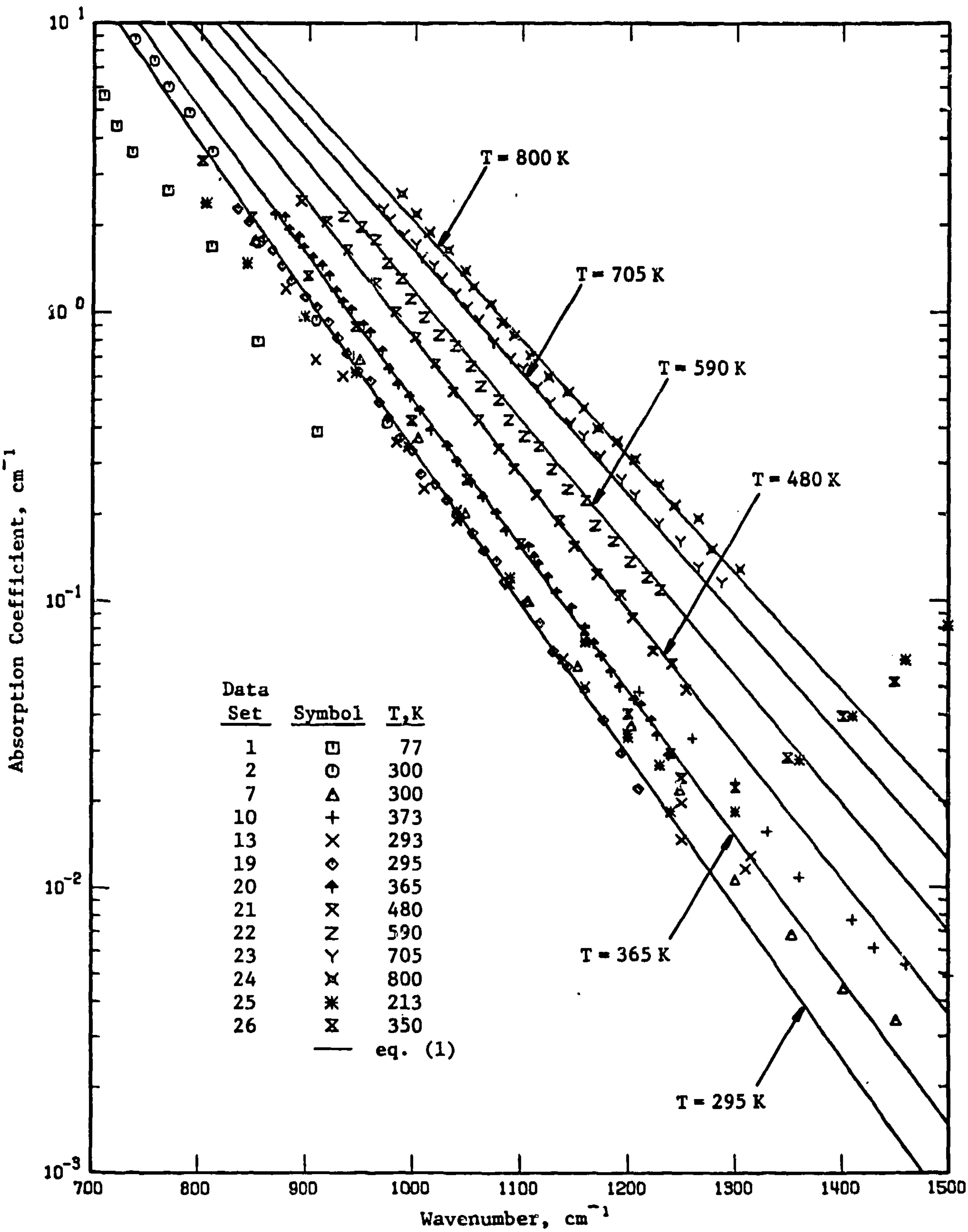

Figure 1. Absorption Coefficient of Strontium Fluoride in the Multiphonon Region (Wavenumber Dependence)

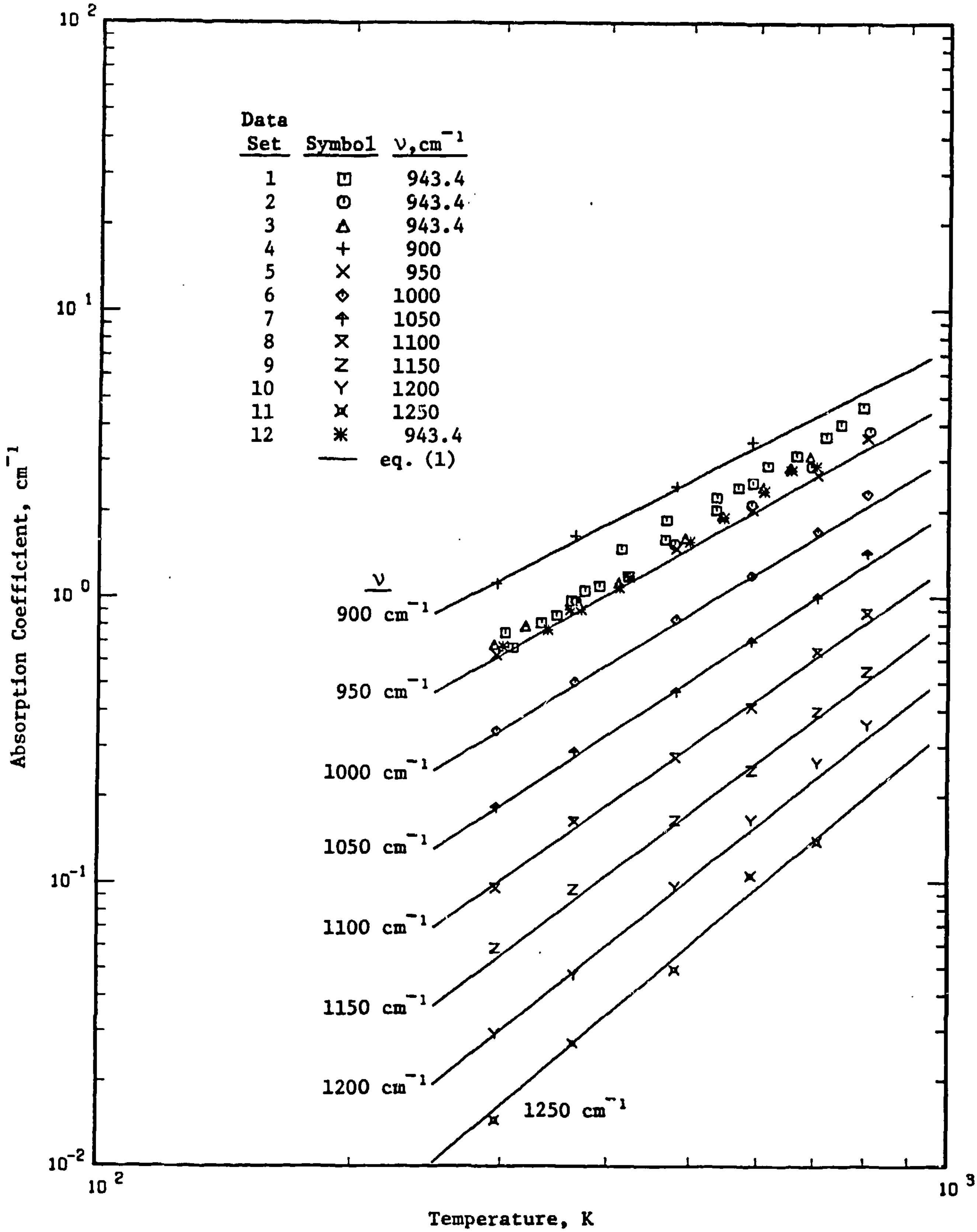

Figure 2. Absorption Coefficient of Strontium Fluoride in the Multiphonon Region (Temperature Dependence).

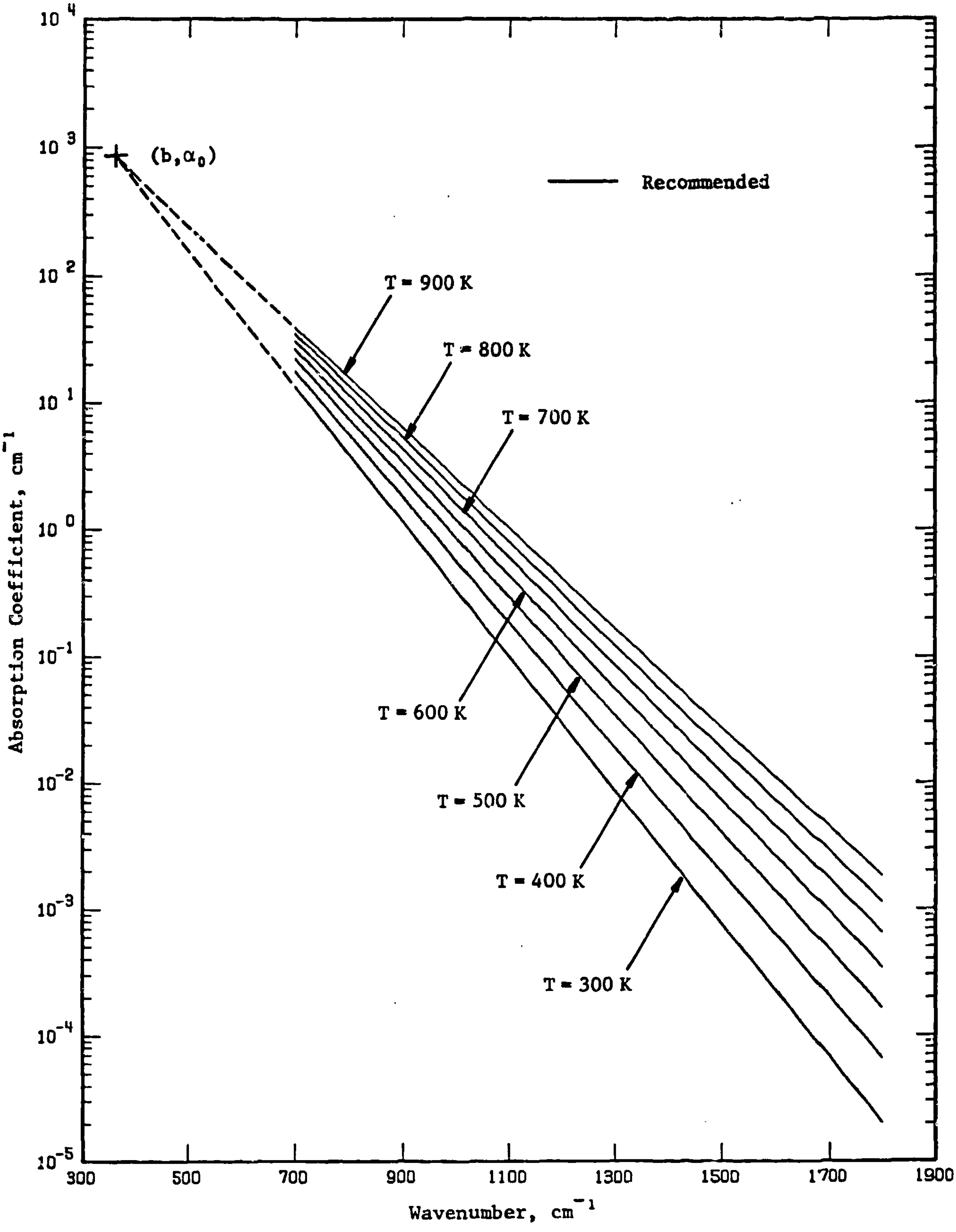

Figure 3. Calculated Absorption Spectra of Strontium Fluoride (Wavenumber Dependence)

PUBLICATIONS RESULTING FROM THE PROJECT

H. H. Li, "Refractive Index of Alkali Halides and Its Wavelength and Temperature Derivatives," J. Phys. Chem. Ref. Data, 5(2), 329-528, 1976.

H. H. Li, "Refractive Index of Alkaline Earth Halides and Its Wavelength and Temperature Derivatives," J. Phys. Chem. Ref. Data, 9(1), 1980.

H. H. Li, "Refractive Index of Silicon and Germanium and Its Wavelength and Temperature Derivatives," J. Phys. Chem. Ref. Data, scheduled to appear in Vol. 9, No. 2, 1980.

H. H. Li, "Absorption Coefficient of Alkali Halides (Part I)," CINDAS Technical Report 54, submitted to AFOSR in March 1979.

H. H. Li, "Absorption Coefficient of Alkali Halides (Part II)," CINDAS Technical Report 55, submitted to AFOSR in July 1979.

H. H. Li, "The Infrared Absorption Coefficient of Alkali Halides," International Journal of Thermophysics, 1(1), 97-134, 1980.

H. H. Li, "Absorption Coefficient of Alkaline Earth Halides" CINDAS Technical Report 57, submitted to AFOSR in April 1980. The contents of the report will be the essential material of a paper of a technical journal after it is approved for release.

PROFESSIONAL PERSONNEL ASSOCIATED WITH THE RESEARCH EFFORT
(May 1, 1979 - April 30, 1980)

During this contractual period, the professional staff members who have contributed to the various tasks of this program are as follows:

Dr. H. H. Li

Dr. T. C. Chi

Dr. Y. S. Touloukian

Mrs. A. F. Furdyna

In addition to the above professional effort, invaluable contribution was made by the professional personnel of the Division of Scientific Documentation at CINDAS in the literature search, data source identification and document procurement.

INTERACTIONS

The director of CINDAS, Dr. Y. S. Touloukian, participated in the following meeting during 1979. He has contacted and discussed the recent development of high-power laser technology with the atendees and gained information useful to this research project.

1. Eleventh Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO, Oct. 30-31, 1979.
2. High Power Laser Optical Components Conference, Boulder, CO, Nov. 1-2, 1979.

Dr. Touloukian has been constantly communicating Dr. A. H. Guenther of the Air Force Weapons Laboratory at Kirtland, New Mexico regarding the subject of optical constants of laser window materials.

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MAGNESIUM FLUORIDE

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

During this period, the work on the absorption coefficient of alkaline earth halides, and germanium and silicon constituted the core activity. The work for alkaline earth halides was finished and a technical report entitled "The Absorption Coefficient of Alkaline Earth Halides" was completed and submitted for inspection and acceptance in April 1980. In this report, experimental material on the absorption coefficients of alkaline earth halides was searched, compiled, and analyzed. It was found that the bulk of available

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data was concentrated to the absorption edges of the main transparent region for the four materials calcium fluoride, strontium fluoride, barium fluoride, and magnesium fluoride. An equation was formulated to best describe the absorption data in the infrared multiphonon region as a function of both frequency and temperature. It was noted that the same type of equation is equally valid for both alkaline earth halides and alkali halides. Although the intrinsic absorption was predicted by the proposed equation, discrepancies might occur in the region where absorption is extremely low and where extrinsic absorptions due to impurities and surface contamination dominate the intrinsic absorption by factors ranging from fractions to multiples of ten. Experience has shown that extrinsic contributions can be reduced through improved crystal growing and surface polishing techniques. The work on the absorption coefficient of silicon and germanium was in progress. It was found that the absorption coefficient is sensitive to the amount and kind of impurities usually being introduced as desirable dopants. Numerous experimental material was made available. Currently the work remained in the stage of data extraction and compilation.